Interfaces in Thermoelectric Materials

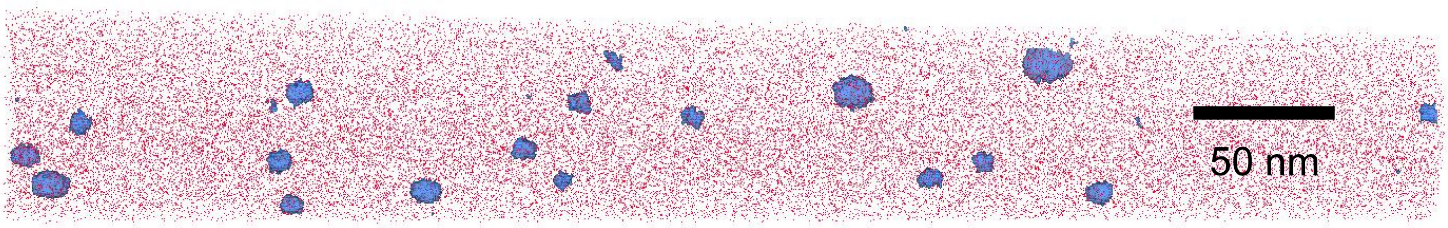

Figure 1: Using advanced microscopy techniques, such as atom probe tomography, Sandia researchers are investigating the mechanisms of nanostructure formation in thermoelectric materials. This example shows the three-dimensional distribution of embedded nanoscale Ag_2Te precipitates in PbTe.

Experiment and theory are providing new insights in nanostructured thermoelectric alloys

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Thermoelectric materials have many applications in the conversion of thermal energy to electrical power and in solid-state cooling. Although thermoelectric devices have found many specialized applications where their high reliability, lack of moving parts, and ability to be scaled to small sizes provide key advantages, the energy conversion efficiencies of these devices remains generally poor. For more wide-spread use of thermoelectrics, the material and device efficiencies will need to be improved dramatically. To address this need, Sandia is building an effort that bridges synthesis, characterization, and thermoelectric measurements, as well as theory and modeling, to harness the potential of nanostructured bulk materials for thermoelectricity.

The energy conversion efficiency of a material is characterized by the thermoelectric figure-of-merit:

$$zT = \frac{\alpha^2 T}{\rho \kappa}$$

Approaches to improve thermoelectric conversion efficiency are driven by the need to maximize the Seebeck coefficient, α, and to balance the competing requirements of low electrical resistivity, ρ, and low thermal conductivity, κ. Interfaces affect each of these properties and can have profound effects when present at the high densities typical of nanomaterials. Recent advances have shown that zT can be enhanced in nanoscale systems by taking advantage of phonon scattering at interfaces to reduce thermal conductivity and quantum confinement and carrier scattering effects to enhance the power factor, α^2/ρ.

Sandia is investigating the formation and stability of interfaces in thermoelectric nanomaterials and how these interfaces control thermal and electronic transport. Advanced microscopic tools, such as atom probe tomography (Figure 1) and electron microscopy, are being used to better understand how embedded nanostructures can form in bulk thermoelectric alloys. For instance, recent work on $AgSbTe_2$, a high-performance thermoelectric material, has clarified the complex phase transformation mechanisms that occur in this system. This material forms fine-scale precipitates of silver telluride (Ag_2Te), which is monoclinic at room temperature. By establishing how these precipitates are oriented (Figure 2), Sandia has shown that the Te-sublattice remains aligned in both the matrix and precipitate phase. Because the monoclinic and high-temperature cubic phases of Ag_2Te differ by only small, local distortions of the crystal lattice, this result suggests a facile transformation path for the Ag_2Te precipitation, requiring only the clustering of excess silver. Given similar observations in PbTe-based materials, it appears that this mechanism is generic to rock-salt structured tellurides.

The experimental work is supported and guided by theory and modeling, where Sandia is developing models to better explain how nanostructures improve thermoelectric properties. Recently, it was shown that nanoscale metallic precipitates can enhance the thermoelectric properties of bulk semiconductors. The idea behind this work is that the presence of a potential well or barrier near the nanoparticle surface causes low-energy electrons to be strongly scattered while allowing high-energy electrons to be unaffected (Figure 3). Thus the nanoparticle/matrix interface serves as an energy filter for electrons. This filtering effect increases the voltage that is generated across a bulk thermoelectric material when a temperature gradient is applied across it (the so-called Seebeck effect). The nanoparticles also play a dual role: because the sound velocity is different in the nanoparticles and the matrix, heat transport through phonons is reduced. This effect, when combined with the electron energy filtering effect, can lead to large enhancement in the energy conversion efficiency.

References:

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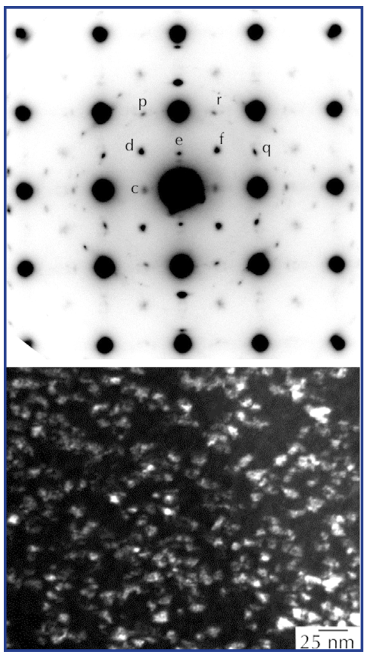

Figure 2: Electron diffraction (top) and dark-field transmission electron micrograph (bottom) showing nanoscale Ag_2Te precipitates in a matrix of thermoelectric $AgSbTe_2$. The orientation results from alignment of the Te sublattices in the two phases.

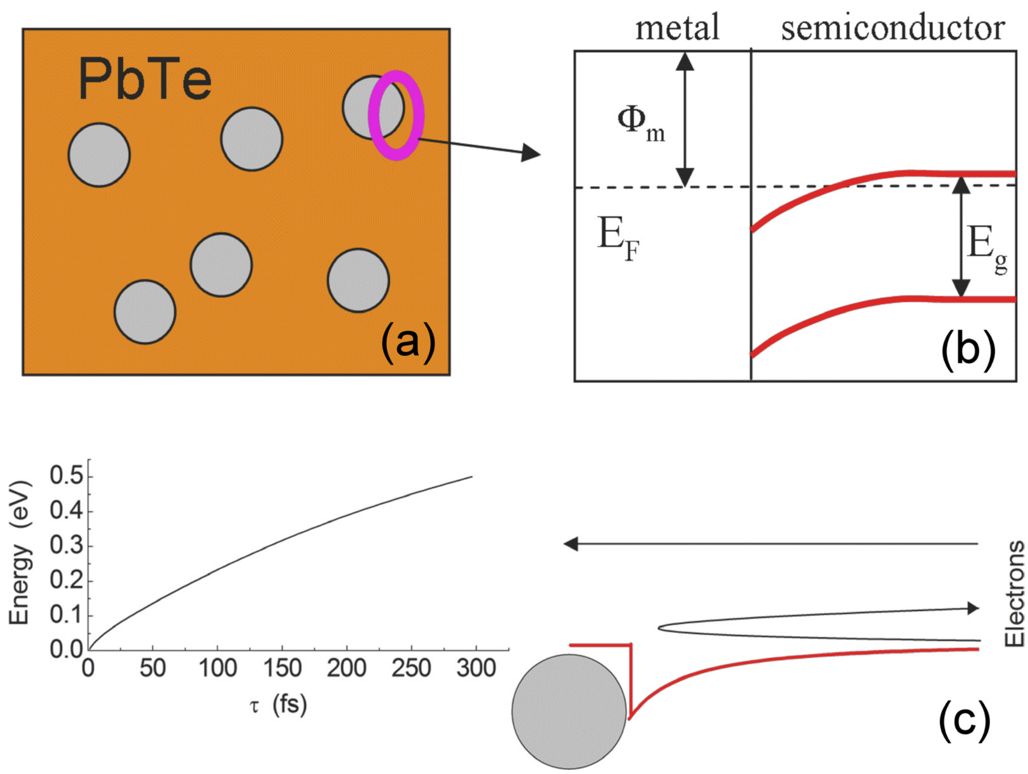

Figure 3: *Panel (a)* Schematic of semiconductor host with metallic nanoinclusions. *Panel (b)* shows an example of the calculated potential and the energy diagram for PbTe with Pb nanoinclusions of radius 1.5 nm. E_F is the Fermi level, E_g is the semiconductor bandgap and Φ_m is the metal work function. *Panel (c)* illustrates the concept of energy filtering: low energy electrons scatter strongly with the potential, but high energy electrons are unaffected. The calculated electronic relaxation time for the potential of panel (b) is also shown.